



Optical properties of internally mixed mineral dust and black carbon

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Perturbations of the optical properties of mineral dust particles by mixing with black carbon: a numerical simulation study

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Abstract

Field observations show that individual aerosol particles are a complex mixture of a wide variety of species, reflecting different sources and physico-chemical transformations. The impacts of individual aerosol morphology and mixing characteristics on the Earth system are not yet fully understood. Here we present a sensitivity study on climate relevant aerosols' optical properties to various approximations. Based on aerosol samples collected in various geographical locations, we have observationally constrained size, morphology and mixing, and accordingly simulated, using the discrete dipole approximation model (DDSCAT), optical properties of 3 aerosols types: (1) bare black carbon (BC) aggregates, (2) bare mineral dust, and (3) an internal mixture of a BC aggregate laying on top of a mineral dust particle, also referred as polluted dust.

DDSCAT predicts optical properties and their spectral dependence consistently with observations for all the studied cases. Predicted values of mass absorption, scattering and extinction coefficients (MAC, MSC, MEC) for bare BC show a weak dependence on the BC aggregate size, while the asymmetry parameter (g) shows the opposites. The simulated optical properties of bare mineral dust present a large variability depending on the modeled dust shape, confirming the limited range of applicability of spheroids over different types and size of mineral dust aerosols, in agreement with previous modeling studies. The polluted dust cases show a strong decrease in MAC values with the increase in dust particle size (for the same BC size), while an increase of the single scattering albedo (SSA). Further, polluted dust particles with radius between 180–300 nm are characterized by a decrease in SSA values compared to bare dust, in agreement with field observations.

This paper demonstrates that observationally constrained DDSCAT simulations allow to better understand the variability of the measured aerosol optical properties in ambient air, and to define benchmarks biases due to different approximations in aerosol parametrization.

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1 Introduction

Black carbon (BC), a distinct type of carbonaceous aerosol particle, is produced by incomplete combustion of fossil and biomass fuels. BC is a strong light absorber and therefore can contribute to atmospheric warming and surface dimming. Estimates of direct BC radiative forcing (DRF) are highly uncertain and range from 0.2 to 1.2 W m⁻² at the top of the atmosphere (TOA) (Bond et al., 2013). Two main sources of DRF uncertainty are: (1) estimates of BC spatial distribution and (2) interaction of BC with electromagnetic waves (EMW) upon emission and after aging in the atmosphere. Realistic modeling of BC spatial distribution relies on proper parametrization of emission, life time and vertical distribution (Samset et al., 2013; Bond et al., 2013), while appropriate modeling of BC interaction with EMW relies on proper parameterization of aerosol shape, chemical composition and state of mixing with other aerosol compounds. Comparison between predicted spatial concentrations of BC from chemical transport models and AERONET measurements, shows consistent biases. In the specific, the fraction of aerosol column (extinction) attributable to absorption, the Aerosol Absorption Optical Depth (AAOD), is generally underestimated by models compared to values retrieved by AERONET (Bond et al., 2013; Koch et al., 2009). The sources of discrepancy are not well understood; In order to estimate BC DRF “consistently” with observations, scaling factors of the order of 2–3, need to be introduced to BC emission estimates to match observed AAOD values. In Koch et al. (2009) BC predictions from the AeroCom model inter-comparison project showed a low model bias for AAOD, but an overestimation of surface and upper BC concentrations at lower latitudes. The authors suggest that most models are underestimating BC absorption and recommend to work on improving estimates of refractive indices, particle size, and optical effects of BC mixing. Many transport models assume BC to be externally mixed with other aerosol compounds, while few models assume that BC is homogeneously internally mixed with other aerosol compounds (Bond and Bergstrom, 2006; Koch et al., 2009). Differences in the representation of the aerosol mixing (i.e., BC with non absorbing

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aerosols) lead to different absorption values, which compared to measurements are too small in the case of external mixing and too high for the case of homogeneous internal mixing. Less often an encapsulation of a spherical and homogeneous absorbing core surrounded by a spherical and homogeneous host material is adopted (core-shell configuration), which gives more realistic magnitudes of absorption (Jacobson, 2014, 2001).

Despite that, not always core-shell configuration can represent the absorption variability in the laboratory and field observations (Adachi et al., 2010; Bueno et al., 2011; Bond et al., 2013; Cappa et al., 2012a, b). The latter might be due to the misrepresentation of the BC particle aggregation and mixing, as shown by more detailed light scattering modeling studies performed by Kahnert (2010b); Scarnato et al. (2013) and Adachi and Buseck (2013). Recent studies show that internal mixing of BC with other aerosol materials in the atmosphere can alter its aggregate shape (Zhang et al., 2008; Xue et al., 2009; Cross et al., 2010; China et al., 2013), absorption of solar radiation (Bueno et al., 2011; Cappa et al., 2012a), and radiative forcing (Adachi et al., 2010; Kahnert et al., 2012). China et al. (2013) and (2014) further, characterized the predominant mixing and morphology types observed with the electron microscopes from samples collected in different locations and for different sources (i.e., biomass burning aerosol and vehicle exhaust) by classifying BC into four main classes (bare BC, inclusions, thinly coated and embedded BC); similar classes were identified by Scarnato et al. (2013) for laboratory generated mixtures of BC and sodium chloride (an aerosol mixture resembling dirty marine aerosol).

Several field campaigns have been showing the occurrence of internal mixing of BC with dust aerosols in the accumulation mode (e.g., Clarke et al., 2004; Liu et al., 2008). During transport and aging in the atmosphere, various processes can result in the formation of multi-component aerosols containing dust, including: (i) heterogeneous chemistry, (ii) adsorption of water vapor on dust particle surfaces, (iii) cloud processing, (iv) coagulation of dust with other aerosol or cloud particles (see Usher et al., 2003). On a global scale, bare dust is estimated to cool the Earth atmosphere. Mineral

example, during the monsoon in Asia (Lau and Kim, 2006) or the African jet streams (Reale et al., 2011).

An accurate parametrization of aerosol optical properties due to variability in morphology and mixing with other aerosol compounds is crucial for a number of disciplines involving not only radiative forcing analysis (Bond et al., 2013) and global and regional aerosol modeling (Samset et al., 2013; Kahnert, 2010b), but also aerosol-cloud interactions, visibility and precipitation forecast (Lau and Kim, 2006) and, further, remote sensing of atmosphere and ocean color (Russell and Heintzenburg, 2000; Durkee et al., 2000; Yoshida et al., 2013).

For example, retrievals of aerosol (and ocean) properties require assumption of: (1) scattering phase function, (2) single scattering albedo (SSA), (3) estimates of ozone absorption and molecular scattering, and (4) for satellite applications, estimates of surface reflectance/albedo. Both orbital and ground based remote sensing techniques use in the analysis of radiometric data a pre-selected library of aerosol types with spherical shape (or in some cases spheroidal shape i.e. for AERONET) and the retrieval algorithm selects an aerosol type based on the best fit to radiance measurements (i.e., Deuze et al., 2001; Hasekamp et al., 2011; Dubovik et al., 2006).

The capability of the satellite and AERONET aerosol global network to provide spatio-temporal distributions of both dust and BC at different spatial scales, relies on how well the aerosol library used in the retrieval “fits” the aerosol mixture in the atmosphere; therefore, it is dependent on the accuracy of the retrieval assumptions on dust and BC optical properties. Therefore, non-sphericity and chemical anisotropy of the particles are sources of potential inaccuracy and biases of data products. These inaccuracies may affect the retrieval of aerosol characteristics, such as refractive index, size, aerosol optical depth, aerosol absorption optical depth, etc. (e.g., Scarnato et al., 2013, and references therein).

In this paper we present an “observationally-constrained” sensitivity study of the optical properties of BC aggregates internally mixed with mineral dust aerosols in the UV–IR spectral range (computationally intensive). The spectral range used in this study is of

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interest for applications in climate modeling, remote sensing of aerosol and ocean properties, and visibility forecast. Computations are performed using the Discrete Dipole approximation (DDA–DDSCAT7.3) (Draine and Flatau, 1994), a technique for modeling particles with complex shape, chains and aggregates with anisotropic mixing. DDSCAT is based on the direct solution of the Maxwell equations without reference to the wave equation, which is usually used in the treatment of light scattering by simple shapes, such as sphere and spheroids. Synthetic particle design for the DDSCAT calculations is based upon single particle electron microscopy of bare BC, bare dust and BC-dust internally mixed particles BC on the surface of a dust particle) collected in the atmosphere from different locations of the globe.

2 Method

2.1 Description of the synthetic particles

2.1.1 Black carbon aggregates

We generate synthetic BC aggregates by aggregation of monomers in random walk (Richard and Davis, 2008; Richard et al., 2011). The synthetic BC aggregates are characterized by a volume equivalent radius (a_{eff} also defined as the radius of a sphere containing all the volume of the particle) between 82–144 nm, a constant monomer diameter of 40 nm and an open-chain like structure. Such values have been observationally constrained after processing electron microscopes images of several aerosol samples collected in field campaigns carried out in different geographical locations, such as California’s Sacramento Valley (during the Carbonaceous Aerosol Radiative Effects Study – CARES), the North Atlantic at the Pico Mountain Observatory on Pico Island in the Azores (PICO), Mexico City, Mexico (Megacity Initiative: Local and Global Research Observations – MILAGRO), Detling, England (Clear air for London – ClearfLo), where internal mixture of BC and mineral dust have been observed.

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The reader should be aware that the morphological characterization of the ambient aerosol is determined by processing 2-D electron microscope images on aerosol particles laying on a substrate and assuming orientational anisotropy over a statistically representative sample. Therefore, as a *minor secondary aspect of the paper*, we investigated the appropriateness of the standard method adopted in literature to estimate a morphological descriptor, such as monomers number, of BC aggregates by performing image processing of two-dimensional (2-D) projections of *synthetic* BC fractal aggregates. Image processing of synthetic BC fractal aggregates allowed, as well, to assure that the synthetic particles have similar 2-D properties to those collected in the atmosphere.

In this paper, we describe the BC aggregate morphology and chain like structure in terms of (1) fractal dimension, porosity (Shen et al., 2008; Scarnato et al., 2013) and convexity (as descriptors of the chain topology), (2) aspect ratio, and (3) roundness.

BC particles can be represented as fractals, where each particle is described as an aggregate with monomers of the same size, approximately obeying the following scaling law

$$N_i = k_0(R_g/r_m)^{D_i} \quad (1)$$

Where N is the number of monomers per aggregate with $i = (\text{estimated}, \text{true})$, R_g is the radius of gyration, r_m is the monomer radius, k_0 is the fractal prefactor, here used a value of $k_0 = 1.6$ (Liu et al., 2008), and D_f is the mass fractal dimension.

R_g is the root-mean-square distance from the center of each monomer to the aggregate center of mass. As often it is difficult to measure N_{true} from 2-D projections, for particles with $D_f < 2$, N_{true} is typically estimated as the ratio of the projected area of the aggregate (A_a) and the mean projected area of a monomer (A_p) in the aggregate (Oh and Sorensen, 1997; Samson et al., 1987).

$$N_i = K_a(A_a/A_p)^\alpha \quad (2)$$

Where α is an empirical projected area exponent and it has a typical value of 1.09, while k_a has a value of 1.15. The sensitivity of N_i on the values of α and k_a has been discussed previously (China et al., 2014).

The aspect ratio (AR) of the fractal aggregate, is defined as the ratio of the major axis (A) to the minor axis (B). Larger values of the aspect ratio indicate a more elongated particle.

$$AR = \frac{A}{B} \quad (3)$$

The roundness of a fractal aggregate, is defined as the ratio of the projected area (A_a) of the particle to the area of a circle with a diameter equal to the maximum length (L_{\max}) of the particle.

$$\text{Roundness} = \frac{4A_a}{\pi L_{\max}^2} \quad (4)$$

The convexity C (also known as solidity), is defined as the ratio of the A_a of the particle and to the area of the smallest convex polygon in which the particle is inscribed (convex hull polygon – CHP). The polygon is calculated based on the boundary enclosing the foreground pixels of a binary image using straight-line segments to each outermost point.

$$C = \frac{A_a}{\text{CHP}} \quad (5)$$

The porosity (P) of the fractal aggregates is defined by Shen et al. (2008)

$$P = 1 - [(\beta_2 + \beta_3 - \beta_1)(\beta_3 + \beta_1 - \beta_2)(\beta_1 + \beta_2 - \beta_3)]^{1/2} \quad (6)$$

where $\beta_i = I_i / (0.4\rho_1 V_1 a_{\text{eff}}^2)$ is a dimensionless quantity, I_i with $i = (1, 2, 3)$ is the moment of inertia tensor, ρ_1 is the density and V_1 the volume of BC aggregates (see Shen et al., 2008).

Morphological descriptors of synthetic BC aggregates are calculated from projected images of 50 randomly particle orientations.

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2.1.2 Mineral dust aerosol

The morphology of suspended mineral dust might take various forms, as natural dust is an aggregate of internally mixed minerals. Different field studies show AR median values ranging between 1.4 and 1.9 (Chou et al., 2006; Clarke et al., 2004; Reid et al., 2003; Kandler et al., 2006; Dubovik et al., 2006; Mishra et al., 2012). In this study, we modeled dust aerosols as spheroids and rectangular prisms with an intermediate aspect ratio of 1.75, which has also been found in CARES, PICO, MILAGRO and ClearfLo field campaigns (see Table 3). We summarize the characteristics of the synthetic/modeled aerosol particles in Table 1. Dust particles with a smaller radius are representative of particle size distribution of long lived distant-transported accumulation-mode airborne dust. The largest radius is representative of the particles size near the dust emission sources.

2.2 Computation of optical properties

We have numerically simulated the optical properties for an ensemble of bare mineral dust, bare open chain-like BC aggregates and internal mixtures of BC and mineral dust, see Table 1. Optical properties of the binary mixtures are modeled using a DDA model (DDSCAT.7.3), see for model details Draine and Flatau (1994, 2010). Numerical simulations have been carried out at the specific spectral channels of the AERONET Cimel radiometer (340, 380, 440, 500, 675, 870, 1020 nm) plus at the 550 nm wavelength for comparison with literature values. Optical properties have been averaged over 1000 random orientations, reference refractive indices are listed in Table 2.

The optical properties discussed in this study are:

1. The mass absorption, scattering and extinction coefficient (MAC, MSC and MEC)

$$\text{MAC} = C_{\text{abs}}/\text{mass} \quad (7)$$

$$\text{MSC} = C_{\text{scat}}/\text{mass} \quad (8)$$

$$\text{MEC} = C_{\text{ext}}/\text{mass} \quad (9)$$

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$$\text{mass} = \rho \frac{4}{3} \pi a_{\text{eff}}^3 = \frac{4}{3} \pi (\rho_1 a_{1,\text{eff}}^3 + \rho_2 a_{2,\text{eff}}^3) \quad (10)$$

where C_{abs} , C_{scat} and C_{ext} indicate the absorption, scattering and extinction cross sections, $\rho_{1,2}$ is the density (index 1 indicates BC and index 2 mineral dust). MAC and MSC are necessary to calculate the effects of mass concentrations simulated by chemical transport models on radiative transfer. MAC and single scattering albedo (SSA) (defined in Eq. 12) are relevant to determine the balance between negative and positive forcing.

2. The aerosol absorption, extinction and scattering Ångström exponents (AAE, EAE, SAE) computed from the slope of the linear fit passing through MAC, MSC and MEC curves (in log–log scale). The AAE and EAE are typically used as indicators of aerosol type and size.

$$\text{AAE} = \frac{-\Delta \log(\text{MAC})}{\Delta \log(\lambda)}; \quad \text{EAE} = \frac{-\Delta \log(\text{MEC})}{\Delta \log(\lambda)}; \quad \text{SAE} = \frac{-\Delta \log(\text{MSC})}{\Delta \log(\lambda)} \quad (11)$$

3. The single scattering albedo (SSA) calculated as:

$$\text{SSA} = C_{\text{scat}}(\lambda) / C_{\text{ext}}(\lambda); \quad (12)$$

where $C_{\text{abs, scat, ext}}(\lambda)$ are defined in Eq. (7).

4. The asymmetry parameter defined as:

$$g = 1/2 \int_0^\pi \cos(\theta) \sin(\theta) P(\theta) d\theta \quad (13)$$

where $P(\theta)$ is the scattering phase function and θ is the scattering angle.

SSA and g are the two fundamental parameters necessary to perform calculations of aerosol radiative properties (e.g., Chylek and Wong, 1995).

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3 Results

3.1 BC internally mixed with dust

We have observed BC internal mixing with suspended mineral dust (BC particles laying on top of dust particles) in various field campaigns. In Fig. 1, we show a composite of SEM images from aerosol samples collected: (a) in an urban location 10 km north of downtown Mexico City (MILAGRO, March 2006), (b) 40 km downwind of the Sacramento urban area in the forested Sierra Nevada foothills, California, USA during CARES (2010), (Zaveri et al., 2012), (c) in a rural site in Detling, UK (ClearfLo, January–February 2012), (d) at Pico Mountain Observatory, Azores Islands (Portugal) in the North Atlantic Ocean (Honrath et al., 2004; Dzepina et al., 2014; China et al., 2015).

The morphological characteristics of the BC and mineral dust particles are summarized in Table 3. The values reported for BC, in Table 3, are in agreement with Adachi et al. (2007).

3.2 Morphological characterization of the synthetic aggregates

In order to ensure that the shape of the synthetic BC aggregates are representative of ambient air samples, we processed the 2-D binary images of the synthetic particles at 50 random orientations. For synthetic aggregates presented in Fig. 2, we have estimated $N_{\text{estimated}}$ values (\pm SD) of 63 (\pm 8) for BL1, 119 (\pm 13) for BL2, 179 (\pm 12) for BL3 and 326 (\pm 46) for BL4. Morphological descriptors for the cases BL1 to BL4 are summarized in Table 4. Aggregates have the same monomer size, similar chain structure, but increasing number of monomers.

The accuracy of $N_{\text{estimated}}$ values after image processing is conditional to two main factors: (1) the number of orientations taken for images processing, and (2) the size of the aggregate. In Fig. 3, we present a comparison of the $N_{\text{estimated}}$ values from the 2-D

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projected images with the actual N_{true} values used for the generation of the synthetic aggregates. The $N_{\text{estimated}}$ well approximate N_{true} within the uncertainties.

3.3 Optical properties of bare BC aggregates

The spectral dependence of mass extinction, absorption and scattering coefficients (MEC, MAC, MSC) is presented in Fig. 4 for an ensemble of synthetic open chain-like aggregates, as described in Table 4 (and with a size parameter $X = 2\pi a_{\text{eff}}/\lambda < 4.5$).

It is well known that bare/uncoated fresh BC absorbs more radiation, than scatters (Bond et al., 2013). Therefore, MAC represents the dominant contributor to the MEC. Bond et al. (2013) report BC MAC values larger than $5 \text{ m}^2 \text{ g}^{-1}$. Predicted values of MEC, MAC and MSC (see Eq. 7) are shown in Fig. 4 for a composite of BC aggregates with similar porosity and monomers size, but increasing numbers of monomers, see Table 4. MAC values are strongly wavelength dependent (see also Moosmueller et al., 1998, and Lack and Langridge, 2013). At 550 nm MAC predicted values, using a BC density (ρ) of 1.8 g cm^{-3} (Bond and Bergstrom, 2006), range between 5.32 and $5.65 \text{ m}^2 \text{ g}^{-1}$ and they are weakly sensitive to the aggregate size. The latter finding is in agreement with the fractal theory by Berry and Percival (1986), which maintains that mass absorption coefficient should not be a strong function of the size, but rather a strong function of the refractive index, physical shape (and mixing) (e.g. Fuller et al., 1999; Liu et al., 2008; Scarnato et al., 2013).

The range of predicted MAC values at 550 nm is in agreement with field measurements by Clarke et al. (2004) and modeled values by Kahnert (2010a) and Kahnert and Devasthale (2011).

However, several studies (e.g., Bond and Bergstrom, 2006; Adachi et al., 2007; Cross et al., 2010) reports larger values. Reasons might be related to different index of refraction or density values, for instance, the values predicted here are lower than the published values at 550 nm by Scarnato et al. (2013) because of differences in the adopted refractive indices. At a wavelength of 550 nm, the refractive index by Chang and Charalampopoulos (1990), adopted in these simulations, have lower real and

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imaginary indices than the value of $1.95-0.79i$ recommended by Bond and Bergstrom (2006) (see Table 2), which was adopted in simulations by Scarnato et al. (2013). In this study, as in Scarnato et al. (2013), we used a BC ρ_1 value of 1.8 g cm^{-3} . If we use a value of ρ_1 equal to 1.4 g cm^{-3} and Chang and Charalampopoulos (1990) refractive index, we find for the cases BL1 to BL4 MAC values at 550 nm of about $7 \text{ m}^2 \text{ g}^{-1}$. As a reminder, the OPAC code uses a density value as low as 1 g cm^{-3} for BC. MSC and SSA values, as shown in Fig. 5, are slightly more sensitive to the aggregates size than the MAC (see also Scarnato et al. (2013) for the SSA dependence on aggregate compactness). SSA values are lower than those predicted by Scarnato et al. (2013), due as well to the differences in the refractive indices used in the simulations. The SSA magnitude and spectral variation presented in this study are both in agreement with laboratory measurements by Sharma et al. (2013). At 550 nm, SSA shows little variability through case BL1 to BL4 with an average value of 0.19 ± 0.02 . Just looking at MAC and MEC, one could argue that the implementation of optical properties of bare BC aggregates in chemical transport and radiative transfer models might be greatly facilitated by the fact that some of the properties of BC aggregates are little sensitive to aggregate size in the UV, VIS and NIR. Such a property would reduce the need for complex parametrizations of BC aggregates optical properties to accurate modeling of the chain structure, and monomer size of the aggregate (see (Liu et al., 2008) for sensitivity to monomer size). This assumption fails when looking at the asymmetry parameter (g) spectral dependency for the cases BL1 to BL4 in Fig. 5b, where g presents a strong sensitivity to the BC aggregate size in the entire spectral range under study. In Fig. 5, DDSCAT predicts the lowest g values for the BL1 case, intermediate values for the case BL4 and higher values for cases BL2 and BL3. For wavelengths longer than 800 nm the differences in g values between the cases BL2, BL3 and BL4 are minimised.

AAE, EAE and SAE values are wavelength dependent, see Table 4 and Scarnato et al. (2013). In the spectral range between 340–1600 nm, AAE values are consistent with observations and theoretical results with values of approximately 1, while in

the spectral range between 400–675 nm AAE values approach 1.2 (in agreement with Lack and Langridge, 2013). The range of values of AAE, EAE and SAE is also fairly consistent with Sharma et al. (2013). For example, we found a SAE average value of 1.79 ± 0.37 , which is in the range of values reported by Sharma et al. (2013) of 1.61 ± 0.05 , and by Gyawali et al. (2012) of 1.88.

3.4 Optical properties of mineral dust

In all the field campaigns presented here, we have found mineral dust particles with jagged surfaces and irregular shape (see Fig. 1). In particular, in Fig. 1a, c, and d dust particles were found to be silica rich and with a plate-like morphology. We found that the DDSCAT predicted optical properties have a large variability depending on the modeled dust shape, despite having the same aspect ratio. In Fig. 6, we present the residual of the $Q_{\text{abs, scat, ext}} = C_{\text{abs, scat, ext}} / \pi a_{\text{eff}}^2$ for an ensemble of spheroids (E1, E2, E3, E4, E5) and rectangular prisms (S1, S2, S3, S4, S5) with $AR = 1.75$. The difference in the $Q_{\text{abs, scat, ext}}$ is small for case E1 and S1, and it is larger (up to about 50 % in the $Q_{\text{ext, scat}}$ at 550 nm) for larger particle sizes (cases S4, S5).

The sensitivity of $Q_{\text{abs, scat, ext}}$ to shape confirms the limited range of applicability of spheroids over different types and size of mineral dust aerosols, in agreement with previous modeling studies Merikallio et al. (2011); Hansell Jr. et al. (2011); Otto et al. (2011). Extended studies on the sensitivity to shape of mineral dust particles optical properties in the UV-NIR can provide useful constrains on the envelope of values to be expected during measurements in ambient air (i.e., Sokolik and Toon, 1999; Hansell Jr. et al., 2011). From Fig. 6, it is also evident that simplifications, in handling mineral dust particle shape, can generate positive (and at times negative) biases in retrieved AOD and opacity, when ellipsoids are adopted in the retrieval and aerosol at the site resemble more the synthetic rectangular prisms/modeled particles. The magnitude of the biases is dependent on the wavelength and size of the particles. For example, if aerosols at the site resemble more the rectangular prism than ellipsoidal shape, large positive biases (up to 50 %) in retrieved AOD can be expected at 550 nm for particles

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with an a_{eff} between 700 and 1000 nm, as mineral dust particles (cases S4, S5) modeled as rectangular prisms have a higher Q_{ext} than ellipsoids (cases E4, E5). No AOD biases should be expected at 550 nm depending on the two shape assumptions for particles smaller than 700 nm. While, an average AOD bias of $15 \pm 7\%$ in the shorter wavelength range (340–500 nm) and $10 \pm 13\%$ for longer wavelengths range (550–1020 nm) should be expected.

3.5 Optical properties of internally mixed BC aggregates

We have modeled a binary internal mixtures of BC aggregate and mineral dust, as visualized in Fig. 7. The BC aggregates is on the surface of the mineral dust particle. Given the plate like structure of Fig. 1a, c and d, we opted to model mineral dust shape as rectangular prisms. The chosen shape does not cover the whole range of variability encountered in ambient air, but for our cases (see Fig. 1), it adds a degree of complexity in the description of mineral dust shape compared to ellipsoids.

In Fig. 8, we present the MAC, MSC, MEC spectral dependence for 3 different aerosol types: (1) an ensemble of bare mineral dust particles with aspect ratio of 1.75 and increasing size (cases S1 through S5), (2) one bare BC aggregate (case BL2), and (3) internal mixtures of the two types (cases BL2S1 through BL2S5, where BL2 is mixed respectively with S1, S2, S3, and S5).

Bare mineral dust aerosols (see cases S1 through S5 in Fig. 8) have low MAC values compared to bare BC aggregates (i.e., case BL2 in Fig. 8) in the UV and NIR region. The MAC values of bare dust are wavelength dependent with predicted larger values in the UV-VIS. Smaller dust particles have higher MAC. DDSCAT predicts for bare/unpolluted dust at 550 nm an MAC average value of $0.13 \pm 0.03 \text{ m}^2 \text{ g}^{-1}$ (\pm SD).

The internally mixed particles (cases BL2S1 through BL2S5, also refereed as polluted dust) have higher MAC values for smaller particles (BL2 has the highest MAC). As expected, DDSCAT predicts higher MAC values for polluted dust than unpolluted/bare dust, with an average MAC value of $0.26 \pm 0.27 \text{ m}^2 \text{ g}^{-1}$ at 550 nm.

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Further, MSC values of bare mineral dust aerosols have a strong variability with size and wavelength. DDSCAT predicts an average MSC value at 550 nm of $2.1 \pm 1.9 \text{ m}^2 \text{ g}^{-1}$ for dust particles ranging in size from 0.18 to 1 micron. When considering just the accumulation mode, with dust size ranging between 0.5 and 1 microns, DDSCAT predicts a smaller MSC average value of $0.8 \pm 0.2 \text{ m}^2 \text{ g}^{-1}$. Fine mode particles compared to coarse mode particles have larger MSC values because smaller particles scatter light more efficiently at visible wavelengths. Hand and Malm (2007), after reviewing 60 studies of ground based observations, report at 550 nm for the fine mode dust an average MSC value of $3.3 \pm 0.6 \text{ m}^2 \text{ g}^{-1}$, while they report in the accumulation mode smaller MSC values of $0.9 \pm 0.8 \text{ m}^2 \text{ g}^{-1}$, in agreement with our study. The MSC of larger dust particles (cases S3, S4, S5) does not show a strong spectral dependence, while the opposite is true for smaller particles (cases S1 and S2), see Fig. 8c. It should be noted that the spectral variability of AOD is used in remote sensing in interpreting aerosol type. For example, mineral dust aerosol is assumed to have a “spectrally flat” AOD, while biomass burning or polluted aerosol usually exhibit a strong wavelength dependence. The spectral dependencies in Fig. 8a demonstrates that small mineral dust aerosol particles and polluted dust have also a strong AOD spectral dependence, those characteristic might be a potential source of classifications of aerosol type, size and amount.

Further, representation of the state of aerosol mixing, whether internal (such cases BL2Si with $i=(1,2,3,5)$) or external (such as cases BL2 plus Si $i=(1,2,3,5)$) might affect the overall optical properties of the aerosols, see Fig. 9. We found that for smaller particles (cases S1, S2, BL2, BL2S1, BL2S2) external and internal mixtures predict similar values of $C_{\text{abs, scat, ext}}$ in the entire spectral range, with ratios respectively of 1.09 ± 0.06 , 0.96 ± 0.05 and 1.02 ± 0.07 . The latter might be due to the combination of small electromagnetic interactions between the BC aggregate and the mineral dust particle and small particles sizes (small size parameter). While, we found larger differences in predicted $C_{\text{ext, abs, scat}}$ values, when BL2 is mixed externally or internally with larger mineral dust particles (cases BL2+S3, BL2+S5, BL2S3 and BL2S5). For those cases, simulations using external mixtures representations give smaller C_{abs} val-

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ues compared to internal mixtures (with average ratio of 0.87 ± 0.30) for wavelengths shorter than 550 nm, while larger values (average ratio of 1.35 ± 0.49) for wavelengths larger than 550 nm. Further, C_{scat} values of external mixtures are smaller than internal mixtures in most of the spectral range studied (and similarly for C_{ext} values) with average ratios of 0.59 ± 0.30 and 0.49 ± 0.27 for wavelength shorter and larger than 550 nm.

The SSA spectral signatures of bare BC (BL2), an ensemble of mineral dust (cases S1 through S5), and internal mixtures of the two aerosol components (BL2S1 through BL2S5) are shown in Fig. 10. Bare mineral dust (case S1 through S5) shows a typical decrease in the SSA magnitude for wavelengths shorter than 500 nm, with SSA values ranging from 0.85 to 0.96 depending on the size of the dust particle, with smaller values attributed to larger particles. The range of values predicted by DDSCAT, in this study, is in agreement with values of 0.7 to 0.97 for Sahara dust reported by Ryder et al. (2013), where the authors attributed variability in measured values to the presence of a significant number of large particles. Further, analyses of the SSA values of Saharan dust from the Aerosol Robotic Network (AERONET) reported averages of 0.95 at 0.67 microns (Dubovik et al., 2002). SSA values of 0.95–0.99 have been reported during the Saharan Dust Experiment (SHADE) and the Dust Outflow and Deposition to the Ocean (DODO) (Tanre et al., 2003; McConnell et al., 2010; Johnson et al., 2008). Osborne et al. (2008) estimated the SSA for pure dust aerosol during the Dust and Biomass-burning Experiment DABEX, (Haywood et al., 2008) to be consistently high (ranging between 0.98–0.99).

For wavelengths shorter than 500 nm, small polluted dust particles (BL2S1–BL2S2) show a stronger decrease in the SSA magnitude compared to unpolluted dust particles (S1–S2), perturbation of dust optical properties of the same order of magnitude was also found in the Aerosol Characterization Experiment (ACE) field campaign (Clarke et al., 2004). While DDSCAT predicts for internally mixed particles larger than 500 nm (BL2S3–BL2S5) an increase of SSA at all wavelengths compared to bare dust particles (S3–S4–S5).

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DDSCAT predicts a wavelength dependent asymmetry parameter g , BC has higher spectral dependence than dust, mostly due to the variation in real part of the BC refractive index with wavelength. DDSCAT predicts at 550 nm higher g values for internally mixed polluted dust than bare mineral dust; larger g values are predicted when modeling an external mixture compared to external mixture, differences can amount up to about 37 % (see Table 6).

4 Conclusions

Microscope images of ambient air aerosol samples collected in various locations of the globe show occurrence of internal mixture of BC aggregates and mineral dust aerosols, see also (Clarke et al., 2004; Haywood et al., 2008). The aerosols shape/morphology and state of mixing, whether internal or external can affect the interaction with EMW and the overall optical properties of the aerosols mixtures, contributing therefore to uncertainty in (1) DRF estimates, (2) validation of chemical transport models with remote sensing measurements, (3) visibility forecast and (4) spatial and temporal distribution of precipitations and their forecast.

In this study, we carried out numerical simulations to investigate on the sensitivity of climate relevant aerosols optical properties to various approximations on aerosol size, shape and state of mixing, and draw benchmarks considerations for climate studies and remote sensing applications. Based on aerosol samples collected in Mexico, England, USA (California) and Portugal, we have observationally constrained morphology and mixing, and modeled optical properties accordingly, of 3 different types of aerosols: (1) bare BC aggregates, (2) bare mineral dust, and (3) an internal mixture of BC and dust particles, also referred as polluted dust.

Optical properties including MAC, MEC, MSC, AAE, EAE, SSA and g were predicted over the spectral range between 340 and 1020 nm using DDSCAT, which applies the discrete dipole approximation (DDA). Specific wavelengths have been selected to match the AERONET nominal channels.

Key results for bare BC aggregates include: (i) a weak MAC dependence on the aggregate size, but stronger MAC dependence on the refractive index, in agreement with Berry and Percival (1986); Liu et al. (2008); Scarnato et al. (2013), (ii) a strong g dependence on aggregate size, (iii) consistence between DDSCAT predicted and observed values of both AAE, EAE, SAE (e.g., Lack and Langridge, 2013) and SSA (and its spectral variability) (Sharma et al., 2013).

Key results for bare mineral dust aerosol include: (i) a strong sensitivity of dust optical properties to shape (DDSCAT predicts at 550 nm an average difference between spheroids and prisms of about 20 % for MEC and MSC, while of about 5 % for the MAC), (ii) consistence between DDSCAT predicted and observed values of MAC, MSC and SSA reported by Hand and Malm (2007); and (iii) a typical decrease in the SSA magnitude for wavelengths shorter than 500 nm (also found to be characteristic of organics and the aerosol mixtures of Sodium Chloride and BC, see also Scarnato et al., 2013; Russell et al., 2010).

Key results for polluted mineral dust, an internal mixture of BC and mineral dust, include: (i) a strong decrease in MAC values with the increase in dust particle size (case BL2S1 presents largest values), while the opposite for SSA values. (ii) A decrease in the SSA magnitude compared to bare dust for smaller dust particle sizes (cases BL2S1 and BL2S2) in agreement with Clarke et al. (2004). Further, (iii) strong differences in predicted magnitude and spectral dependence of $C_{\text{abs, scat, ext}}$ when mixing a BC aggregate (case BL2) externally or internally with large mineral dust particles (cases S3, S5, BL2S3, BL2S5).

With this study, we demonstrated the importance of (i) characterizing and defining micro-physical properties, such as morphology/shape and mixing of different aerosol types collected in ambient air, (ii) estimating optical properties accordingly to observations, and (iii) in defining eventual benchmarks errors due to use of approximations in shape and mixing. More studies are needed to assess the abundance of polluted dust particles in the atmosphere. In fact, the occurrence of such configuration is currently highly uncertain and might strongly depend on source and transport regions. Account-

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ing for changes in optical properties induced by mixing as well as of the abundance of mixed particles, might be critical not only for calculating the relevance of such particles on regional radiative forcing, but also to understand biases in remote sensing techniques and to explore the potential of such techniques in remotely detecting mixed particles cases.

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Table 1. Characteristics of the modeled aerosol particles. Dust particles have an aspect ratio (AR) of 1.75 in all cases.

Aerosol Type	Legend	Target shape in study	a_{eff} [nm]
Dust	S1, E1	Rectangular prism, Ellipsoid	180
	S2, E2	Rectangular prism, Ellipsoid	280
	S3, E3	Rectangular prism, Ellipsoid	500
	S4, E4	Rectangular prism, Ellipsoid	700
	S5, E5	Rectangular prism, Ellipsoid	1000
Black Carbon	BL1	BC aggregate with 70 monomers	80
	BL2	BC aggregate with 100 monomers	100
	BL3	BC aggregate with 200 monomers	120
	BL4	BC aggregate with 300 monomers	140
Mineral dust and BC	BL2S1	Rectangular prism mixed with BC (BL2 + S1)	190
	BL2S2	Rectangular prism mixed with BC (BL2 + S2)	290
	BL2S3	Rectangular prism mixed with BC (BL2 + S3)	503
	BL2S5	Rectangular prism mixed with BC (BL2 + S5)	1010

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Table 2. References of the wavelength dependent refractive indices and density values used for BC and mineral dust. Chang and Charalampopoulos (1990) values at about 550 nm are $1.77 - 0.63i$, which are lower than the value of $1.95 - 0.79i$ suggested by Bond and Bergstrom (2006).

Aerosol Compounds	Reference Refractive Indices	Density [g cm^{-3}]
Black Carbon	Chang and Charalampopoulos (1990)	1.8
Mineral Dust	Wagner et al. (2012)	2.6

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Table 3. Synthesis of morphological descriptors for BC and mineral dust aerosol particles sampled in various field campaigns.

Aerosol Type	Projected Area Equivalent Radius [nm]	AR	r_m [nm]
Dust	250–810	1.08–1.75	
Black Carbon	90–140	1.39–1.98	34–49

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Table 4. Morphological characterization of synthetic BC particle.

Case	$2r_m$ [nm]	N_{true}	a_{eff} [nm]	P	Convexity	D_f	AR	Roundness
BL1	40	70	82	0.86	0.65 ± 0.05	1.98 ± 0.09	1.36 ± 0.17	0.43 ± 0.08
BL2	40	100	100	0.92	0.66 ± 0.05	1.95 ± 0.05	1.46 ± 0.22	0.38 ± 0.04
BL3	40	200	126	0.89	0.63 ± 0.04	2.03 ± 0.05	1.34 ± 0.16	0.44 ± 0.05
BL4	40	300	144	0.90	0.60 ± 0.06	1.95 ± 0.07	1.74 ± 0.36	0.34 ± 0.07

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Table 5. Summary of optical properties predicted by DDSCAT for bare BC aggregates at 550 nm. AAE and EAE have been calculated in different wavelength ranges: (a) 340–1000, (b) 400–675 and (c) 340–1600 nm – (spectral range not shown in Fig. 4). MAC and MEC values are provided at 550 nm. MSC values have not been included in the table, as they can be calculated by the difference between MEC and MAC values.

Case	AAE a (b) (c)	EAE a (b) (c)	SAE a (b) (c)	MAC(550) m^2g^{-1}	MEC(550) m^2g^{-1}	SSA(550)	g(550)
BL1	1.26 (1.24) (1.04)	1.36 (1.33) (1.15)	1.49 (1.32) (1.82)	5.60	6.80	0.17	0.42
BL2	1.24 (1.23) (1.11)	1.38 (1.41) (1.26)	1.58 (2.20) (2.32)	5.72	6.93	0.17	0.60
BL3	1.18 (1.21) (1.05)	1.29 (1.32) (1.18)	1.33 (1.73) (1.90)	5.38	6.94	0.22	0.73
BL4	1.18 (1.18) (1.04)	1.27 (1.35) (1.15)	1.60 (1.92) (1.70)	5.53	6.98	0.20	0.76

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Table 6. Summary of simulated optical properties for Mineral Dust and internal mixtures with BC aggregates. AAE and EAE have been calculated in two different wavelength ranges: (a) 340–1020 and (b) 400–675 nm.

Case	a_{eff} [nm]	AAE a (b)	EAE a (b)	SEA a (b)	MAC (550) m^2g^{-1}	MEC (550) m^2g^{-1}	SSA (550 nm)	g (550 nm)
S1	180	3.92 (5.68)	2.8 (2.62)	2.75 (2.37)	0.14	3.70	0.96	0.75
S2	284	2.90 (4.16)	0.22 (0.95)	0.25 (1.56)	0.17	5.10	0.96	0.81
S3	500	2.15 (3.70)	0.65 (0.80)	1.17 (0.18)	0.12	0.82	0.85	0.60
S4	700	1.89 (3.13)	0.10 (0.47)	0.69 (1.5)	0.13	1.32	0.90	0.79
S5	1000	1.53 (2.60)	0.18 (0.26)	0.74 (0.60)	0.10	0.70	0.86	0.81
BL2S1	190	1.84 (2.39)	2.20 (2.23)	2.46 (2.19)	0.64	3.97	0.84	0.77
BL2S2	289	2.00 (2.48)	1.08 (0.14)	0.27 (1.63)	0.27	5.00	0.94	0.81
BL2S3	503	2.10 (3.17)	0.19 (0.93)	0.34 (2.68)	0.13	2.19	0.94	0.79
BL2S5	1010	2.68 (4.20)	0.45 (0.97)	0.92 (1.48)	0.09	2.82	0.96	0.88

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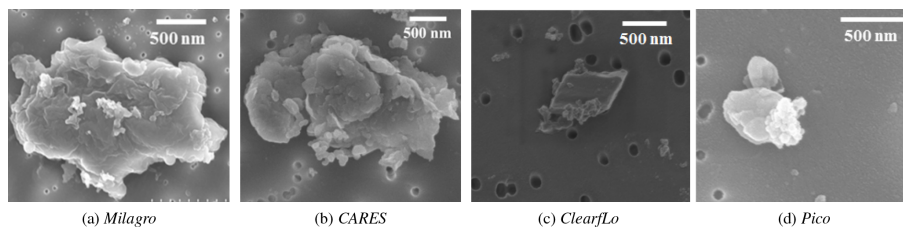


Figure 1. SEM images of internally mixed mineral dust and BC particles observed during various field campaigns, **(a)** Mexico City, Mexico (Megacity Initiative: Local and Global Research Observations (MILAGRO) 2006); **(b)** a silicon-rich dust particle internally mixed with BC, Sacramento, California, USA (CARES, 2012); **(c)** a complex internal mixture of multiple aerosol components Detling, UK (Clear Air for London (ClearLo), 2012); and **(d)** a dust particle with plate-like structure (clay-mineral) from Pico Mountain Observatory, Pico island, Azores (Portugal) in the North Atlantic Ocean (2012).

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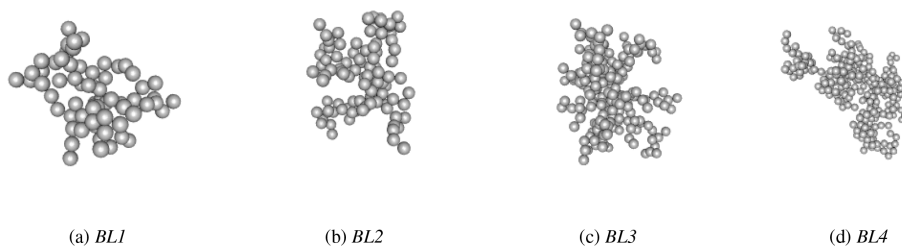


Figure 2. Representation of four BC aggregates (not in scale) with increasing number of monomers (see Table 4). In all cases the monomers radius is 20 nm.

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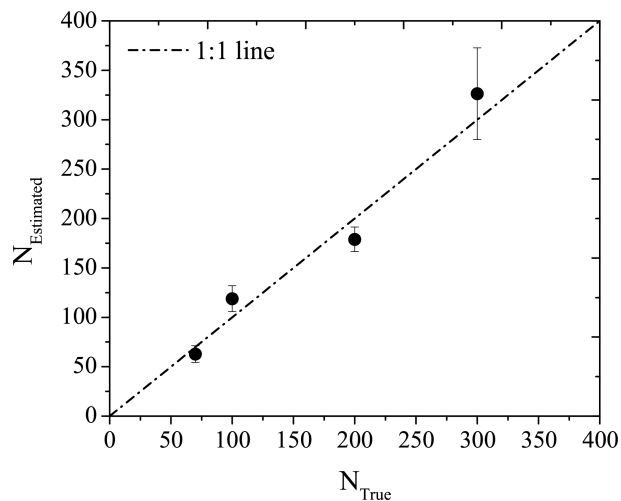


Figure 3. Comparison between the actual monomers number (N_{true}) of the synthetic BC aggregates vs. the estimated monomers number ($N_{estimated}$) after image processing of 2-D projections of 50 random aggregate orientations, the error bars represent the SD.

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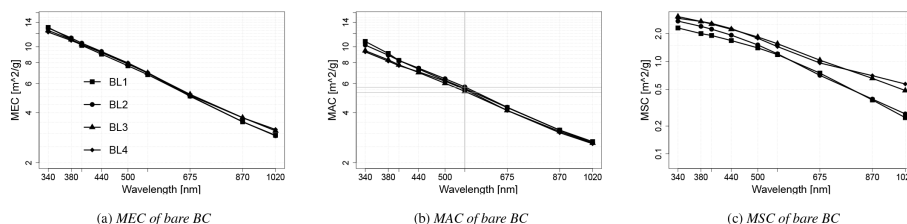


Figure 4. (a) MEC (b) MAC (c) MSC spectral dependence (in log–log scale) for an ensemble of BC aggregates, as described in Table 1. All the computed optical values are averaged over 1000 particle orientations. Dark grey lines underlines MAC values at 550 nm. Note that the y scale is different in the 3 plots.

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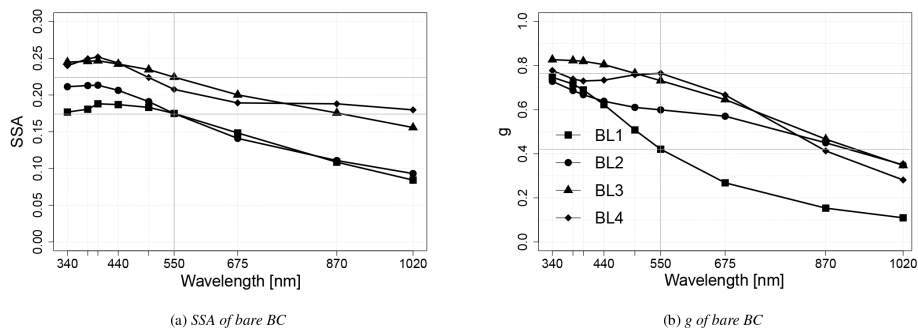


Figure 5. (a) SSA and (b) g spectral dependence for bare BC aggregates (cases BL1, BL2, BL3, BL4). All the computed optical values are averaged over 1000 particle orientations.

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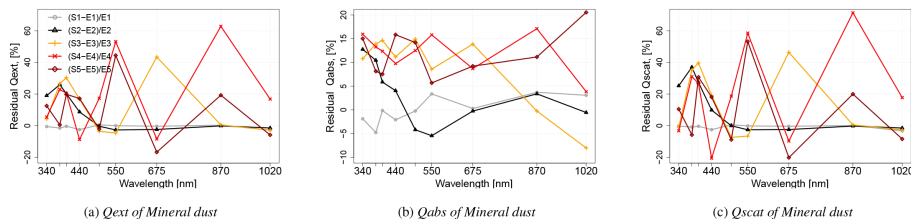


Figure 6. Differences in percentage between extinction, absorption and scattering efficiency for spheroids vs. rectangular prisms.

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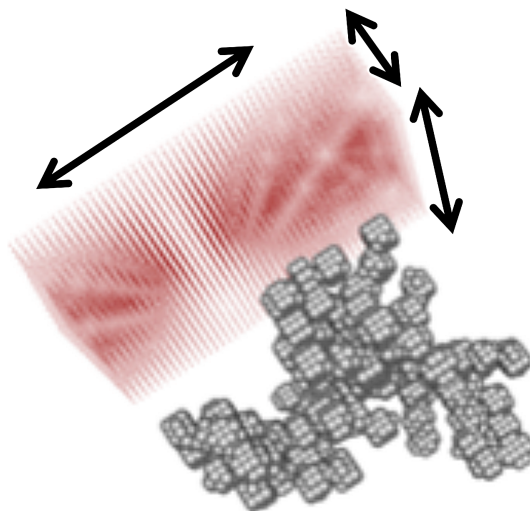


Figure 7. Visual representation of polluted dust, as an internal mixture of BC and mineral dust. The shape of the particle is represented by an array of coordinates (small dots or spheres), to which is associated a dipole moment. Brown dots represent the dust particle dipoles, while grey small spheres represent the dipoles of the BC aggregate. The cases BL2S1, BL2S2, BL2S3, BL2S5 have BL2, respectively, on the surface of S1, S2, S3 and S5. Arrows show that sides of the rectangular prism can vary keeping aspect ratio constant to a value of 1.75.

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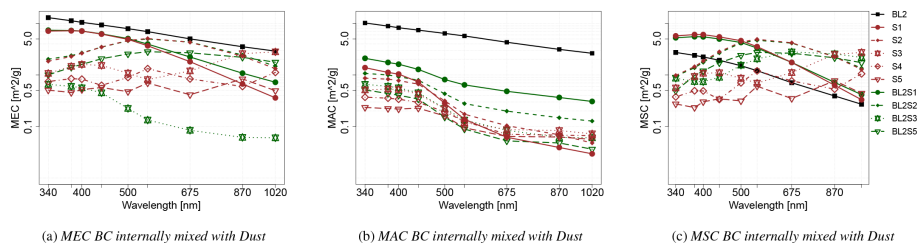


Figure 8. (a) MEC (b) MAC (c) MSC spectral dependency for a BC aggregate (BL2) bare mineral dust (S1–S5) and internal mixtures (BL2S1–BL2S5), see Fig. 7. MEC, MAC, MSC are normalized *by total mass* of the internally mixed particle, see Table 2. All the computed optical values are averaged over 1000 particle orientations.

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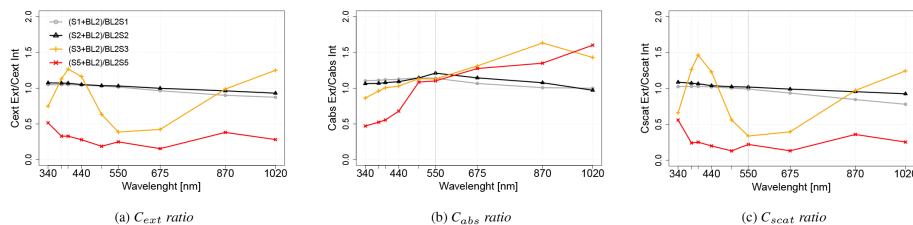


Figure 9. (a) C_{ext} , (b) C_{abs} and (c) C_{scat} ratios of external vs. internal mixtures of BC and mineral dust aerosols.

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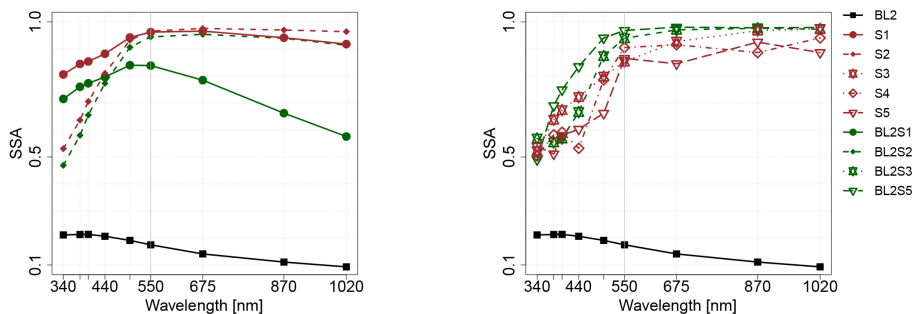
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Interactive Discussion



Optical properties of internally mixed mineral dust and black carbon

B. V. Scarnato et al.



(a) SSA spectral dependence for smaller particles size

(b) SSA spectral dependence for larger particle size

Figure 10. SSA for different particle sizes in the accumulation mode: **(a)** finer particles **(b)** larger particles.

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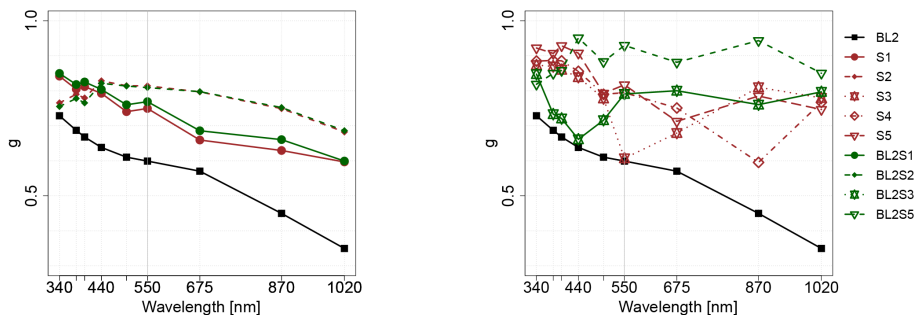
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